## Spin transitions in a small Si quantum dot

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We have studied the magnetic field dependence of the ground state energies in a small Si quantum dot. At low fields the first five electrons are added in a spin-up – spin-down sequence minimizing the total spin. This sequence does not hold for larger number of electrons in the dot. At high fields the dot undergoes transitions between states with different spins driven entirely by Zeeman energy. We identify some features that can be attributed to transitions between different spin configurations preserving the total spin of the dot. For a few peaks we observed large linear shifts that correspond to the change of the spin of the dot by 3/2. Such a change requires that an electron in the dot flips its spin during every tunneling event.

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The spin degree of freedom is an essential part of mesoscopic physics. Quite often the knowledge of the spin advances our understanding of electron-electron interactions, which can reveal themselves in a non-trivial spin configuration of a mesoscopic system. From this perspective, quantum dots can be regarded as model systems for the study of spin-related phenomena because they contain just a few electrons and different coupling parameters can be tuned almost independently [1]. Within the simplest model of non-interacting electrons each additional electron is added into the dot to the next singleparticle energy level (there is also a constant energy associated with the charging of the environment). The spin is accounted for by allowing two electrons to fill the same single-particle energy level, thus the total spin of the system should alternate between s=0 and s=1/2. There are several ways to determine the spin of the dot. The most direct way is by studying the Kondo effect [2–4]. If the spin of the ground state s > 0, the Coulomb blockade is lifted in the corresponding conduction valley at low temperatures. Indeed, valleys with Kondoenhanced conductivity were found to alternate with the regular Coulomb blockade valleys of vanishing conductivity. However, if the dot is weakly coupled to the leads the Kondo temperature can be too small to be achieved experimentally. In such dots individual energy levels are sharp and spin of the tunneling electron can be determined from the Zeeman shift of the energy level. The shift of consecutive peaks has been shown to alternate with  $\pm g^* \mu_B B$  in small Al clusters [5] and carbon nanotubes [6], supporting the alternating spin filling of the dot (here  $g^*$  is the effective g-factor and  $\mu_B$  is the Bohr magneton). In lateral semiconductor quantum dots with a large number of electrons the Zeeman shift is masked by much larger orbital effects, and direct determination of the spin is a formidable task. Peaks fluctuate as a function of B, reflecting the orbital shift of the levels. An indirect information about the spin can be obtained from

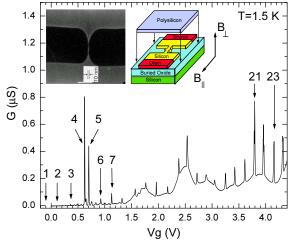


FIG. 1. Conductance as a function of gate voltage measured with  $V_{ac}=100~\mu\mathrm{V}$  at  $T=1.5~\mathrm{K}$  and B=0. Peaks are numbered in sequence starting from the entrance of the first electron into the dot (peak 1). The first two peaks are not resolved at zero bias but their positions can be determined from the high-bias spectroscopy. In the inset a schematic and a scanning electron micrograph of the sample are shown.

the comparison of such "magnetic fingerprints", in order to find whether the two consecutive electrons fill the same energy level. The underlying assumption here is that the addition of an electron does not change the spectrum of the dot significantly. Using this method, strong deviations from the alternating spin filling has been reported [7]. The most dramatic example of a non-alternating spin filling is the polarization of small vertical dots due to exchange interactions, similar to the Hund's rule in atomic physics [8].

The application of a magnetic field alters the spin configuration. Energy levels shift differently with magnetic field and cross each other. In the vicinity of such a magnetically-induced level crossing exchange interactions may lift the degeneracy by favoring the formation of a

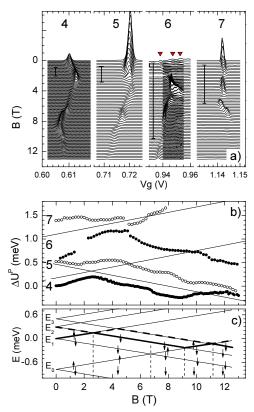


FIG. 2. a) Evolution of four consecutive peaks as a function of B||I. Conductance was measured at 200 mK with  $V_{ac}=50~\mu V$ . Individual traces are offset linearly with B and bars are 1  $\mu S$  scales. In b) peak shifts  $\Delta U^p(B) = [V_g^p(B) - V_g^p(0)]/\alpha$  are plotted for the same four peaks. The zero-field positions are arbitrarily offset. Points are omitted if peak conductance < 0.01  $\mu S$ . Peak 6 is comprised of three peaks at B < 2 T [marked with triangles in a)] and only the lowest-energy branch is shown. Solid lines have a slope of 0.058 meV/T  $(1/2g^*\mu_B$  for  $g^*=2$ ). c) Schematic evolution of single-particle energy levels, assuming that B-dependence enters only through the Zeeman energy. Thin lines have slopes  $\pm 1/2g^*\mu_B B$ . Thick solid and dashed lines follow energies of the 4-th and 5-th electrons. Spins of the four lowest states are indicated by small arrows.

triplet state [9]. A sufficiently large field gradually polarizes the dot by collapsing all electrons into the lowest Landau level [10,11].

In this Letter we examine the electron transport through a small Si quantum dot. Unlike the previously studied semiconductor dots, in our dot the field-dependent shift of energy levels is dominated by the Zeeman energy rather than by orbital effects. Thus, we can measure the spin of the dot directly for ground states with different number of electrons, starting from one. We also study the evolution of the total spin as a function of the magnetic field. We find that the field dependence of energy levels consists of several linear segments with different slopes. Comparison with a simple model for non-interacting electrons allows us to identify a set of

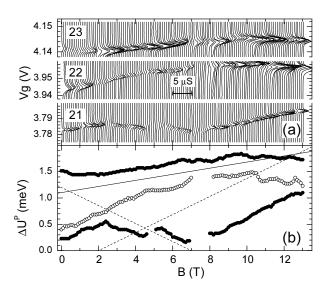


FIG. 3. a) Evolution of peaks 21, 22 and 23 as a function of  $B \perp I$ . Conductance was measured at T=60 mK using  $V_{dc}=20~\mu V$ . All three data sets have the same scale. In b) peak shifts are plotted for the same three peaks, similar to Fig. 2. Solid and dashed lines have slopes 1/2 and  $3/2g^*\mu_B$  respectively.

spin transitions for the first few ground states. In addition to the singlet-triplet and triplet-polarized transitions there are some features in the spectra, which we attribute to transitions between different realizations of the triplet state. A detailed analysis reveals some deviation from the model that hints for the importance of the underlying interactions. As the number of electrons increases, the spectra become more complicated. For example, tunneling of an electron into the dot can change the total spin of the other electrons in the dot. Such a tunneling process is beyond the scope of a model of non-interacting electrons.

The measurements were performed on a small Si quantum dot fabricated from a silicon-on-insulator wafer. The dot resides inside a narrow bridge patterned from the top Si layer (see inset in Fig. 1). A 50 nm thick layer of thermal oxide is grown around the bridge followed by a poly-Si gate. The fabrication steps have been described previously [12]. Gate capacitance is estimated to be 0.8-1.0 aF, the total capacitance  $C \approx 15$  aF and the charging energy  $U_c = e/C \approx 10$  meV. Spacing between excited levels  $\delta \sim 1-4$  meV, measured using non-zero bias spectroscopy, is comparable to the charging energy and is consistent with the lithographical size of the dot  $l \approx \sqrt{\hbar/m^*\delta} \approx 100-190 \text{ Å}$ . The gate voltage – to – energy conversion coefficient, measured from both non-zero bias spectroscopy and T-dependent scaling of the peak width, is  $\alpha \approx 14$  mV/meV. The sample was studied in three separate cooldowns and the reported phenomena were found to be insensitive to redistribution of background charges, thus reflecting intrinsic properties of the

dot.

At high temperatures, T > 120 K, the device exhibits regular metal-oxide-semiconductor field effect transistor (MOSFET) characteristics with a threshold gate voltage of  $V_{th} \approx -0.2$  V. At T < 100 K the Coulomb blockade emerges and the conductance oscillates as a function of the gate voltage  $V_g$  for  $V_g > V_{th}$ . A representative trace of the conductance G as a function of  $V_g$  at T = 1.5 K is shown in Fig. 1. There is a series of sharp peaks, spaced by 150-200 mV. The peaks, corresponding to the entrance of the first two electrons, cannot be reliably measured, but their positions can be determined from high bias spectroscopy. Commonly for this type of devices, the sample has a parallel conducting channel [13], which exhibits Coulomb blockade at  $V_q < 1.2$ V with peaks separated by 60 mV. At  $V_g > 1.2$  V the extra channel has finite conductance with some broad features as a function of  $V_q$ . Coulomb blockade peaks, originated from the lithographical dot, are not broadened at high  $V_q$ , thus electrical transport through the dot and the parallel channel are decoupled. Charging of remote impurities (which can be accomplished by wide gate voltage scans  $\Delta V_g > 3$  V) changes positions of the extra peaks at  $V_q < 1.2$  V and the value of the background conductance at  $V_g > 1.2 \text{ V}$  without altering the position and amplitude of the main peaks.

We studied the peak positions  $V_g^p$  as a function of magnetic field B for the first 30 peaks. For  $V_g < 0.4$  V (the first three peaks), electron density in the contacts is low and the contacts are spin polarized by a moderate magnetic field. For  $V_g > 0.4$  V ( $N \ge 4$ ) both spin subbands in the contacts are occupied within the experimental range of 0 < B < 13 T. Thus, the Fermi energy  $E_F$  is field-independent and the peak shift reflects only the field dependence of the energy levels in the dot (mobility of the two-dimensional gas is low,  $\approx 300 \text{ cm}^2/\text{V} \cdot \text{s}$  at 4.2 K, and there is no measurable modulation of  $E_F$  due to Subnikov-de-Haas oscillations for B up to 13 T).

The evolution of several peaks with B is shown in Fig. 2a (peaks 4-7) and in Fig. 3a (peaks 21-23). Clearly,  $V_q^p$ and the peak amplitudes  $G^p$  change non-monotonically with B. Analysis of "magnetic fingerprints" reveals that there is no apparent pairing of the neighboring peaks within the first 30 peaks. In fact, we observed an unexpected tripling of the peaks: two bunches of peaks have similar "magnetic fingerprints" for three consecutive peaks (13,14,15 and 16,17,18, not shown in the figures). The measurements were repeated for two different orientations of B, defined in the inset in Fig. 1. We found that  $V_q^p$  is insensitive to the direction of the magnetic field: aligning B with the current direction  $(B_{||}, \text{ in-}$ plane) or perpendicular to the plane of the sample  $(B_{\perp})$ does not change  $V_q^p$  significantly. The dot is lithographically asymmetric and the orbital effects are expected to depend on the field direction. Thus, we conclude that in our small dot the B-dependence of  $V_q^p$  is dominated by spin effects. This conclusion is also supported by the observation that, in the range of B when the contacts are fully spin-polarized, the  $V_g^p$  for peaks 1-3 does not depend on B at all.

Unlike  $V_g^p$ , the peak amplitude  $G^p$  depends on the direction of the magnetic field. The  $G^p$  reflects the tunneling probability and depends exponentially on the overlap of wavefunctions in the dot and in the contacts. As such,  $G^p$  is sensitive to a particular configuration of the wavefunction within the dot, and redistribution of the wavefunction due to small orbital effects can result in a significant change of  $G^p$ .

What physics is behind the field-dependence of the peak position? At zero bias  $V_g^p$  is determined by the degeneracy condition that the electrochemical potentials for the ground states with N-1 and N electrons in the dot are equal. Provided that the Fermi energy in the contacts is independent of the magnetic field, a shift of the N-th peak with B reflects the relative change of the ground state energies  $\Delta U_N^p(B) = \Delta U(N,B) - \Delta U(N-1,B)$ , where U(N,B) is the energy of the ground state of N electrons in magnetic field B and  $\Delta U(N, B) = U(N, B)$ U(N,0). In the absence of spin-orbit interactions (which is the case for the bulk Si) the total energy can be separated into spin and orbital terms. The spin term includes Zeeman energy  $s(N)g^*\mu_B B$  where s(N) is the total spin of the ground state with N electrons and  $g^*$  is the effective q-factor. Thus, the Zeeman-related peak shift is  $[s(N) - s(N-1)]g^*\mu_B B = (\pm n \pm 1/2)g^*\mu_B B$ , where the spin  $\pm 1/2$  is carried by the tunneling electron and  $n = 0, 1, 2, \dots$  is the number of electrons in the dot that flip their spins upon the tunneling event. In the simplest case of no interactions peaks should shift linearly with B by  $\pm 1/2q^*\mu_B B$ .

Experimentally, peaks do not shift linearly with B. Instead,  $dV_q^p/dB$  changes both its value and sign as B is varied from 0 to 13 T. For a quantitative analysis, peak positions are extracted from G vs  $V_g$  scans, and the peak shifts  $\Delta U^p(B) = [V_q^p(B) - V_q^p(0)]/\alpha$  are plotted as a function of B in Figs. 2b and 3b. The curves are offset for clarity. For a comparison, lines with slopes  $\pm 1/2g^*\mu_B$ for  $g^* = 2$  are also shown (solid lines). First, let us focus on the low-field (B < 2 T) region. Peaks 4 and 5 shift linearly with B and the corresponding slopes are + and  $-1/2g^*\mu_B$ . In the same low-field region the preceding peaks 2 and 3 also shift with + and  $-1/2g^*\mu_B$ slopes correspondingly. Thus, at low fields the ground states with up to 5 electrons in the dot have the lowest spin configuration and the dot is filled in a spin-down spin-up sequence  $\downarrow\uparrow\downarrow\uparrow\downarrow$  (in the order the levels are filled). Such a filling sequence requires that the valley degeneracy is lifted and two electrons with different spins can occupy the same energy level.

This simple picture of alternating filling does not hold for N>5 even at low fields. At B<2 T peak 6 consists of three peaks separated by  $\approx 0.5$  meV at zero field, none

of which shifts with  $1/2g^*\mu_B B$  (the zero-field positions of the three peaks are marked by triangles in Fig. 2a). The slope of the lowest-energy branch is close to  $3/2g^*\mu_B$ , the other two branches have small negative slopes. The shift of the next, the 7-th, peak has a positive slope, while the lowest-spin arrangement for a dot with 7 electrons should have negative Zeeman energy. We conclude that the ground state with 6 electrons is spontaneously polarized and the total spin s(6) > 1/2. Transitions between ground states that involve a change in spin by  $\Delta s > 1/2$ have low probability and the corresponding peaks are expected to be suppressed (so-called spin blockade [14]). Indeed, the overall conductance of peak 6 is strongly suppressed and, presumably, the appearance of several branches can be explained by the instability of the polarized state.

The low-field spin configuration is not preserved at high magnetic fields. For peak 4,  $dV_a^p/dB$  changes sign from positive to negative at B = 2.5 T, back to positive at B = 9 T, and, again, to negative at  $B \approx 12$ T. The spin of the tunneling electron changes from being  $+1/2 \rightarrow -1/2 \rightarrow +1/2 \rightarrow -1/2$ . The corresponding spin transitions of the ground state can be understood from a simple model for non-interacting electrons. Let us consider four single-particle levels  $E_i$ , as shown in Fig. 2c. Each level is spin-degenerate at zero field and splits into two levels  $E_i \pm 1/2g^*\mu_B B$  for B > 0. In the absence of interactions position of the N-th peak is determined by  $U(N,B)-U(N-1,B)=\sum_k^N E(k,B)-\sum_k^{N-1} E(k,B)=E(N,B)$ , where E(k,B) is the energy of the k-th electron, including the Zeeman contribution. E(4, B) is outlined by the thick solid line in Fig. 2c. Qualitatively, E(4,B) captures the main features of  $V_q^p$  vs. B for the 4-th peak and the kinks can be attributed to the corresponding level crossings. Each level crossing results in a change of the spin configuration and the ground state of 4 electrons undergoes spin transitions as a function of B:  $\downarrow\uparrow\downarrow\uparrow\rightarrow\downarrow\uparrow\downarrow\downarrow\rightarrow\downarrow\downarrow\uparrow\downarrow\rightarrow\downarrow\downarrow\downarrow\uparrow$  (the regions with different spin configurations are separated by dashed vertical lines in Fig. 2c). The first transition is singlet-triplet and the last transition is triplet-spin polarized. There are two intermediate transitions within the triplet state which change the spin configuration within the dot without changing the total spin. At  $B \approx 7$  T the spin configuration of the ground state with 4 electrons changes without reversing the spin of the tunneling electron; such a transition does not change the sign of  $dV_q^p/dB$ . In the absence of interactions there should be no corresponding kink. The second transition flips the spin of the tunneling electron and of an electron in the dot simultaneously, preserving s(4) = -1 but changing the sign of  $dV_a^p/dB$ .

The model, described above, also reproduces the features of peak 5 for B < 7 T (dashed line in Fig. 2c). However, there are some important discrepancies, which cannot be understood within this model of non-interacting electrons. First of all, we cannot describe the evolution of

N>5 peaks within this model. Second, each level crossing should result in a pair of upward–downward kinks in two neighboring peaks at the same value of B. Clearly, kinks in  $V_g^p(B)$  for peaks 4 and 5 near 2 T are shifted by  $\approx 0.5$  T. The most notable deviation from this simple model of level crossing is shown in Fig. 3b, where upward kinks at 2.3 T and 5.3 T in  $V_g^p(B)$  for peak 21 have no downward counterparts in  $V_g^p(B)$  for peak 22. Third, we have to assume a small single-particle level spacing of  $\approx 0.3$  meV to fit the positions of the observed spin transitions. From non-zero bias spectroscopy, as well as from the statistics of the zero-bias peak spacing, we estimate that excited levels are separated by 1-4 meV.

For most of the peaks  $|dV_a^p/dB| \approx 1/2g^*\mu_B$ . However, there are a few peaks that shift much faster with magnetic field. In Fig. 3b  $\Delta U^p(B)$  for peaks 21 and 22 have linear segments with a slope  $\approx 3/2q^*\mu_B B$ . Remarkably, the shift of peak 21 has such a large slope in the whole range 0 < B < 13 T, although its sign changes four times. We can rule out enhancement of the g-factor because i) there are segments in the neighboring peak 23 with the slope  $1/2g^*\mu_B$  (assuming  $g^*=2$ ), and ii) it is known that interactions renormalize  $g^*$  at low electron densities in Si-MOSFETs but  $g^*$  approaches the bulk value of 2 as the density increases [15]. Thus, the total spin of the dot changes by s(N) - s(N-1) = 3/2. A change of the spin by more than 1/2 means that at least one electron in the dot should flip its spin (3/2 = 1+1/2 = 2-1/2) upon the tunneling of an electron. We want to stress the difference with the spin transitions discussed above: there, the total spin of the dot changes as a function of B, but it is fixed for any particular B. In order to change the total spin by 3/2 an electron in the dot has to flip its spin during the tunneling event. In the absence of spin-orbit interactions such a flip is forbidden unless some other spin scattering mechanism is considered. As we mentioned earlier, the absence of an efficient spin scattering should result in a spin blockade with the corresponding suppression of the peak amplitude. Experimentally, there is no apparent suppression of peaks 21 and 22, which have the  $3/2g^*\mu_B$ slopes, compared to the amplitude of peak 23, which has the regular slope of  $1/2g^*\mu_B$ .

To summarize, we have analyzed the field dependence of ground state energies in a small Si quantum dot. The dot is in a new regime where the B-dependence of the energy levels is dominated by the Zeeman energy. There are distinctive features in the data which we attribute to the transitions between different spin configurations of the dot. For the state with 4 electrons in the dot we identified five different spin configurations, including three with the same total spin s=-1. Some peaks have large shift as a function of magnetic field which requires the total spin of the dot to be changed by  $\Delta s > 1/2$  upon the tunneling of an electron. Surprisingly, we found that such peaks are not necessarily suppressed.

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- For a review, see Mesoscopic Electron Transport, Vol. 345 of NATO ASI Series, edited by L. P. Kouwenhoven, L. L. Sohn and G. Schön (Kluwer, London, 1997).
- [2] D. Goldhaber-Gordon, H. Shtrikman, D. Mahalu, D. Abusch-Magler, U. Meirav, and M. A. Kastner, Nature 391, 156 (1998).
- [3] S. M. Cronenwett, T. H. Oosterkamp, and L. P. Kouwenhoven, Science 281, 540 (1998).
- [4] J. Schmid, J. Weis, K. Eberl, and K. v. Klitzing, Physica B 256, 182 (1998).
- [5] D. C. Ralph, C. T. Black, and M. Tinkham, Phys. Rev. Lett. 78, 4087 (1997).
- [6] D. H. Cobden, M. Bockrath, P. L. McEuen, A. G. Rinzler, and R. E. Smalley, Phys. Rev. Lett. 81, 681 (1998).
- [7] D.R. Stewart, D. Sprinzak, C.M Marcus, C.I. Duruoz, and J.S. Harris Jr., Science 278, 1784 (1997).
- [8] S. Tarucha, D. G. Austing, T. Honda, R. J. Van der Hage, and L. P. Kouwenhoven, Phys. Rev. Lett. 77, 3613 (1996).
- [9] W.G. Van der Wiel, T.H. Oosterkamp, J.W. Janssen, L.P. Kouwenhoven, D.G. Austing, T. Honda, and S. Tarucha, Phys. B 256-258, 173 (1998).
- [10] P.L. McEuen, E.B. Foxman, U. Meirav, M.A. Kastner, Y. Meir, N.S. Wingreen, and S.J. Wind, Phys. Rev. Lett. 66, 1926 (1991).
- [11] R.C. Ashoori, Nature 379, 413 (1996).
- [12] E. Leobandung, L. Guo, Y. Wang, and S. Y. Chou, Appl. Phys. Lett. 67, 938 (1995).
- [13] L. P. Rokhinson, L. J. Guo, S. Y. Chou, and D. C. Tsui, Appl. Phys. Lett. 76, 1591 (2000).
- [14] D. Weinmann, W. Häusler, and B. Kramer, Phys. Rev. Lett. 74, 984 (1995).
- [15] T. Ando, A. B. Fowler, and F. Stern, Rev. of Mod. Phys. 54, 437 (1982).